

# Europäisches Patentamt European Patent Office Office européen des brevets



(11) EP 0 786 925 B1

(12)

# **EUROPEAN PATENT SPECIFICATION**

(45) Date of publication and mention of the grant of the patent:19.06.2002 Bulletin 2002/25

(51) Int Cl.7: **H05B 33/14**, C09K 11/06

(21) Application number: 97200088.9

(22) Date of filing: 13.01.1997

# (54) White light-emitting electroluminescent devices

Weiss-ausstrahlende organische Elektrolumineszenzvorrichtungen Dispositifs électroluminescents émettant de la lumière blanche

(84) Designated Contracting States: **DE FR GB** 

(30) Priority: 26.01.1996 US 592830

- (43) Date of publication of application: 30.07.1997 Bulletin 1997/31
- (73) Proprietor: EASTMAN KODAK COMPANY Rochester, New York 14650 (US)
- (72) Inventors:
  - Shi, Jianmin, c/o Eastman Kodak Company Rochester, New York 14650-2201 (US)
  - Tang, Ching Wan, c/o Eastman Kodak Company Rochester, New York 14650-2201 (US)
- (74) Representative: Wagner, Karl H., Dipt.-Ing. WAGNER & GEYER
  Patentanwälte
  Gewürzmühlstrasse 5
  80538 München (DE)

(56) References cited:

US-A- 4 769 292 US-A- 5 189 029 US-A- 5 187 288 US-A- 5 405 709

- PROCEEDINGS OF THE ELECTROCHEMICAL SOCIETY FALL 1995 MEETING. EXTENDED ABSTRACTS, 8 - 13 October 1995, CHICAGO, USA, pages 1316-1317, XP002035633 RH JORDAN ET AL: "White and coloured organic electroluminescent devices for backlights"
- JOURNAL OF APPLIED PHYSICS, vol. 80, no. 12, 15 December 1996, pages 6954-6964, XP002035634 A DODABALAPUR: "Physics and applications of organic microcavity light emitting diodes"
- JOURNAL OF APPLIED PHYSICS, vol. 65, no. 9, 1 May 1989, pages 3610-3616, XP000571884 TANG C W ET AL: "ELECTROLUMINESCENCE OF DOPED ORGANIC THIN FILMS"
- OPTICS LETTERS, vol. 19, no. 20, 15 October 1994, pages 1630-1632, XP000445444
   PARTRIDGE W P ET AL: "PERFORMANCE OF PYRROMETHENE 580 AND 597 IN A COMMERCIAL ND:YAG- PUMPED DYE-LASER SYSTEM"

:P 0 786 925 B1

Note: Within nine months from the publication of the mention of the grant of the European patent, any person may give notice to the European Patent Office of opposition to the European patent granted. Notice of opposition shall be filed in a written reasoned statement. It shall not be deemed to have been filed until the opposition fee has been paid. (Art. 99(1) European Patent Convention).

#### Description

5

20

45

55

# **FIELD OF THE INVENTION**

[0001] This invention relates to organic electroluminescent (EL) devices. More specifically, the invention relates to organic electroluminescent devices which emit white light from a current conducting organic layer.

# **BACKGROUND OF THE INVENTION**

10 [0002] The organic electroluminescent devices which emit white light from a current conducting organic layer have very important applications. The applications of such a device include paper-thin light sources, a backlight for liquid crystal display, and full color displays achieved by combining the emitters with micropatterned color filters. The following patents and publications disclose the preparation of organic EL devices, capable of emitting white light, comprising a hole transporting layer and an organic luminescent layer, and interposed between a pair of electrodes.

**[0003]** Sato in JP 07,142,169 discloses an organic electroluminescent device, capable of emitting white light, is made by staking a blue light emitting layer next to the hole transporting layer and followed by a green light emitting layer having a region containing a red fluorescent dye.

[0004] Kido et al., in Science, Vol. 267, p. 1332, (1995), also in Appl. Phys. Lett. Vol. 64, p. 815, (1994), report a white light-emitting organic electroluminescent device. In this device, three emitter layers with different carrier transport properties, each emitting blue, green, or red light, are used to generate white light.

[0005] Littman et al. in U.S. Patent No. 5,405,709 discloses another emitting organic electroluminescent device which is capable of emitting white light in response to hole-electron recombination and comprises a fluorescent material and a mixed ligand aluminum chelate.

[0006] Tokailin et al. in U.S. Patent No. 5,126,214 taught an electroluminescent element comprising an electroluminescent layer which emits a blue light and a fluorescent layer spaced from the electroluminescent layer. The fluorescent layer absorbs the blue light generated by the electroluminescent layer and fluoresces in a visible light range from bluish green to red. The disclosed element is capable of producing white light.

[0007] However, these EL devices require the use of multiple layers of organic luminescent materials. In order to produce white light, each luminescent layer is necessarily doped with one or more fluorescent dyes. Thus, the fabrication process is complicated.

Furthermore, the emission color may vary, depending on the drive conditions because small changes in voltage may cause electron-hole recombination to take place in different layers producing color variations.

[0008] The related improvement in organic EL devices have been disclosed in U.S. Patent Nos: 5,151,629; 5,150,006; 5,141,671; 5,073,446; 5,061,569; 5,059,862; 5,059,861; 5,047,687; 4,950,950; 4,769,292, 5,104,740; 5,227,252; 5,256,945; 5,069,975, and 5,122,711.

[0009] Jordan et al., in "Proceedings of the Electrochemical Society", fall 1995 meeeting, extended abstract, 8 - 13 October 1995, Chicago, USA, pages 1316-1317, XP002035633, "White and colored organic electroluminescent devices for backlights", report thin-film organic electroluminescent (EL) devices having, in a first approach, a thin layer of a blue-emitting species, 2-naphthyl-4,5-bis(4-methoxyphenyl)-1,3-oxazole (denoted NAPOXA), sandwiched between TAD (bis(triphenyl)diamine, hole transporter) and AlQ (tris(8-hydroxyquinoline)aluminum, electron transporter) to produce white-light emission. In one example of this first approach the organic dye DCM 1 was added to AlQ to broaden the spectrum. In another approach disclosed in this document a portion of an ALQ-layer of an electroluminiscent device is doped with the organic dye pyrromethene (PM 580).

# SUMMARY OF THE INVENTION

[0010] It is an object of this invention to provide a simple electroluminescent device which is capable of emitting white light efficiently.

[0011] In accordance with the invention an electroluminescent device is provided as set forth in claim 1. Preferred embodiments are disclosed in the dependent claim.

[0012] Upon electric excitation by either a DC or AC voltage source, luminescence is produced from this host-guest luminescent layer. Depending on the concentration of the guest molecule in the host, the color of the electroluminescence varies from the blue-green fluorescence of the pure host solid to the red fluorescence of the guest molecule. By selecting an appropriate guest concentration in the host, combination of these two emissions is produced, resulting in white electroluminescence.

#### **ADVANTAGES**

[0013] An advantage of the present invention is that the EL device structure is simple, using a single luminescent layer to produce white light. In particular, the materials of this invention when used as guest dopants in a host material provide a unique combination for producing a white EL device. It is also a feature that a single guest component in the host material can be effectively employed.

[0014] Another advantage is that this white EL device is efficient and stable and should be useful in display and lighting applications.

#### BRIEF DESCRIPTION OF THE DRAWINGS

#### [0015]

10

15

20

FIG. 1 is a schematic cross-sectional view of an electroluminescent device in accordance with the invention. For convenience of illustration, it will be understood that various layers are not to scale since other than the glass layer, they are in the submicron range.

FIG. 2 is a schematic cross-sectional view of an alternative construction of an electroluminescent device in accordance with the invention. For convenience of illustration, it will be understood that various layers are not to scale since other than the glass layer, they are in the submicron range.

#### **DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS**

[0016] Referring now to FIG. 1, an electroluminescent (EL) device 100 is shown. The device is formed on a glass substrate 102. This glass substrate can be borosilicate or soda lime. On the glass substrate 102 there is provided a layer 104 which forms the anode of the device 100. The anode layer typically can be formed from indium tin oxide. On the anode, in sequence, there are provided the following layers, hole injection layer 110, hole transport layer 112, luminescent layer 114 and electron transport layer 116. The layers 110, 112, 114 and 116 all comprise the organic electroluminescent medium 106. On the medium 106 or the electron transport layer 116, is formed the cathode 108. The anode and cathode are connected to an external AC or DC power source 120 conductors 122 and 124, respectively. The power source can be pulsed or continuous wave (CW).

[0017] The device 100 can be viewed as a diode which is forward biased. Under these conditions injection of hole 130 (positive charge carrier) from anode 104 occurs into the lower organic layer, as schematically shown in FIG. 1, while electron (negative charge carrier) are injected into the upper organic layer, as schematically shown at 140, into the luminescent medium. The injected holes and electrons each migrate toward the oppositely charged electrode, as shown by the arrows 132 and 142, respectively. This results in hole-electron recombination. When a migrating electron drops from its conduction band to a valence band in filling a hole, energy is released as light. Hence the organic luminescent medium forms between the electrodes a luminescence zone receiving mobile charge carriers from each electrode. Depending upon the choice of alternative constructions, the released light can be emitted from the organic luminescent material through the anode, through the cathode, or through any combination of the foregoing.

[0018] An alternative construction of the EL device is shown in FIG. 2. In this structure, the hole-injection layer 110 and the electron-injection layer 116 of EL device 100 are omitted. The EL performance based on this simplified structure would still be functional provided the electrode contacts can adequately inject charge carriers into the EL medium. This means that the potential barrier between the anode 104 and the hole-transport layer 112 is sufficiently low such that hole injection from the anode to the hole-transport layer is relatively unimpeded when the device is biased with a low voltage. Likewise, the potential barrier between the cathode 108 and the luminescent layer 114 is sufficiently low such that the electron injection from the cathode to the luminescent layer is also unimpeded. It is understood that in this structure the luminescent layer is capable of electron transport as well as electron-hole recombination is necessary for the production of electroluminescence.

[0019] Other alternative constructions of the EL device based on FIG. 1 are possible. For instance, one such construction would omit only the hole-injection layer 110, but retain all the other layers. Another construction would omit only the electron injection layer 116, but retain all other layers. The criteria for selecting one of these alternative construction is based on a combination of factors, such as the injection properties of the electrode contacts, the ionization potentials of the individual layers in contact with the electrodes as well as the transport characteristics of the individual organic layer comprising the EL medium.

[0020] The hole injection layer 110 of EL device 100 contains a porphyrinic compound. A porphyrinic compound is any compound, natural or synthetic, which is derived from or includes a porphyrin structure, including porphine itself. Any of the prophyrinic compounds disclosed by Adler, U.S. Patent No. 3,935,031 or Tang U.S. Patent No. 4,356,429 can be employed.

[0021] Preferred porphyrinic compounds are those of structural formula (III):

$$T^{1}$$
 $T^{2}$ 
 $T^{2}$ 
 $T^{3}$ 
 $T^{4}$ 
 $T^{2}$ 
 $T^{2}$ 
 $T^{2}$ 

(III)

wherein

10

15

20

25

30

35

40

45

Q is N or C-R;

M is a metal, metal oxide, or metal halide;

R is hydrogen, alkyl, aralkyl, aryl, or alkaryl; and

 $T^1$  and  $T^2$  represent hydrogen or together complete a unsaturated six member ring, which can include substituents, such as alkyl or halogen. Preferred six membered rings are those formed of carbon, sulfur, and nitrogen ring atoms. Preferred alkyl moieties contain from about 1 to 6 carbon atoms while phenyl constitutes a preferred aryl moiety.

[0022] In an alternative preferred form the porphyrinic compounds differ from those of structural formula (III) by substitution of two hydrogens for the metal atom, as indicated by formula (IV):

50 (IV)

[0023] Highly preferred examples of useful porphyrinic compounds are metal free phthalocyanines and metal containing phthalocyanines. While the porphyrinic compounds in general and the phthalocyanines in particular can contain any metal, the metal preferably has a positive valence of two or higher. Exemplary preferred metals are cobalt, magnesium, zinc, palladium, nickel, and, particularly, copper, lead, and platinum.

[0024] Illustrative of useful porphyrinic compounds are the following:

Prophine

1,10,15,20-tetraphenyl-21H,23H-porphine copper (II)

1,10,15,20-tetrapheyl-21H,23H-porphine zinc (II)

Copper phthlocyanine

Chromium phthalocyanine fluoride

[0025] The hole transporting layer of the organic EL device contains at least one hole transporting aromatic tertiary amine, where the latter is understood to be a compound containing at least one trivalent nitrogen atom that is bonded only to carbon atoms, at least one of which is a member of an aromatic ring. In one form the aromatic tertiary amine can be an arylamine, such as a monarylamine, diarylamine, triarylamine, or a polymeric arylamine. Exemplary monomeric triarylamines are illustrated by Klupfel et al. U.S. Patent No. 3,180,730. Other suitable triarylamines substituted with vinyl or vinyl radicals and/or containing at least one active hydrogen containing group are disclosed by Brantley et al. U.S. Patent Nos. 3,567,450 and 3,658,520.

[0026] Another class of aromatic tertiary amines are those which include at least two aromatic tertiary amine moieties. Such compounds include those represented by structural formula (V).

Q1 \_ \_ O2

(V)

wherein

5

15

20

25

30

35

40

45

50

55

Q1 and Q2 are independently aromatic tertiary amine moieties, and

G is a linking group such as an arylene, cycloalkylene, or alkylene group of a carbon to carbon bond.

[0027] A preferred class of triarylamines satisfying structural formula (V) and containing two triarylamine moieties are those satisfying structural formula (VI):

R² | R¹---C--R³ | R⁴

(VI)

where

 $R^1$  and  $R^2$  each independently represents a hydrogen atom, an aryl group, or an alkyl group or  $R^1$  and  $R^2$  together represent the atoms completing a cycloalkyl group, and

R<sup>3</sup> and R<sup>4</sup> each independently represents an aryl group which is in turn substituted with a diaryl substituted amino group, as indicated by structural formula (VII):

 $-N_{R^6}^{R^5}$ 

(VII)

wherein R<sup>5</sup> R<sup>6</sup> are independently selected anyl groups.

[0028] Another preferred class of aromatic tertiary amines are tetraaryldiamines. Preferred tetraaryldiamines include

two diarylamino groups, such as indicated by formula (VIII), linked through an arylene group:

$$R^7$$
  $N$   $Are$   $n$   $N$   $R^8$ 

(VIII)

wherein

5

10

15

20

25

30

40

45

50

55

Are is an arylene group, n is an integer of from 1 to 4, and Ar, R<sup>7</sup>, R<sup>8</sup>, and R<sup>9</sup> are independently selected aryl groups.

[0029] The various alkyl, alkylene, aryl, and arylene moieties of the foregoing structural formulae (V), (VI), (VIII), can each in turn be substituted. Typical substituents including alkyl groups, alkoxy groups, aryl groups, aryloxy groups, and halogen such as fluoride, chloride, and bromide. The various alkyl and alkylene moieties typically contain from about 1 to 6 carbon atoms. The cycloalkyl moieties can contain from 3 to about 10 carbon atoms, but typically contain five, six, or seven ring carbon atoms e.g., cyclopentyl, cyclohexyl, and cycloheptyl ring structures. The aryl and arylene moieties are preferably phenyl and phenylene moieties wherein

Are is an arylene group, n is an integer of from 1 to 4, and Ar, R<sup>7</sup>, R<sup>8</sup>, and R<sup>9</sup> are independently selected aryl groups.

[0030] The various alkyl, alkylene, aryl, and arylene moieties of the foregoing structural formulae (V), (VI), (VIII), can each in turn be substituted. Typical substituents including alkyl groups, alkoxy groups, aryl groups, aryloxy groups, and halogen such as fluoride, chloride, and bromide. The various alkyl and alkylene moieties typically contain from about 1 to 6 carbon atoms. The cycloalkyl moieties can contain from 3 to about 10 carbon atoms, but typically contain five, six, or seven ring carbon atoms-e.g., cyclopentyl, cyclohexyl, and cycloheptyl ring structures. The aryl and arylene moieties are preferably phenyl and phenylene moieties.

[0031] Illustrative of useful hole transport compounds are the following:

$$\begin{array}{c|c} C_2H_5 \\ \hline \\ C_2H_5 \end{array} N \begin{array}{c} C_2H_5 \\ \hline \\ C_2H_5 \end{array}$$

[0032] The luminescent layer emitting white light in device 100 comprises of a host organic material uniformly doped with a small amount of a guest material.

[0033] It is desired to blend with minor amounts of each of one or more fluorescent materials capable of emitting broad band red light and one or more fluorescent materials capable of emitting broad band blue light, the concentrations of each of the materials being selected to produce white emitted light.

[0034] The guest materials of luminescent layer of device 100 contains a fluorescent compound represented by structural formula I:

 $\gamma$ 

5

10

40

50

55

wherein R¹-R³, which may be the same or different, are hydrogen, halogen, or alkyl, alkoxy, alkenyl, cycloalkyl, arylalkyl, acyl, wherein the alkyl portions each contain fewer than 24 carbons, or aryl heteroaryl, alone or in combination.

[0035] This class of fluorescent compounds is known in its use as fluorescent probes because of its high quantum efficiency of fluorescence and other optical properties. For reference, see U.S. Patent No. 5,326,692 and literature cited therein. Particularly useful in organic EL application of the present invention are the following specific compounds.

4.

5

G8

15

20 . G9

30

35

G10

50

45

55

G11

N. j. K.

N. jc.Nt.

F F

S F S

5 10 G12

15

20

25

30

45

55

[0036] The host materials of luminescent layer of device 100 contains the compounds that emit blue green electroluminescence. The host compound is a mixed ligand aluminum chelate, specifically a bis(RS -8-quinolinolato) (phenolato)aluminum(III) chelate of formula II,

$$(R^S - Q - 1)_2 - AI - O - L$$

wherein Q in each occurrence represents a substituted 8-quinolinolato ligand, RS represents an 8-quionolinolato ring substituent chosen to block sterically the attachment of more than two substituted 8-quinolinolato ligands to the aluminum atoms, O-L is a phenolato ligand, and L is a hydrocarbon group that includes an aryl moiety.

[0037] The following constitute specific examples of preferred mixed ligand aluminum chelates useful for the practice of the present invention; other suitable materials are shown in columns 12-17 of U.S. Patent No. 5,150,006:

11

5 H3

H4 H4

10

20

25 H5 20

H6 NC NC AI O AI O 2

45 H7 F<sub>3</sub>C AI O AI O O

Æ.

35

40

50

55

[0038] These above classes of host materials all produce blue-green fluorescence in a pure solid with a high quantum efficiency.

For white EL emission, portion of the electroluminescence is produced by the host material, and the other portion is necessarily produced by the red emitting guest material. Therefore, in construction the white EL emitting layer, it is important to select an appropriate range of concentrations of the guest molecule in the host matrix. Too high a concentration of guest molecule would produce an undesirable red hue from the guest, whereas too low a concentration would produce an equally undesirable blue-green hue from the host. It is found that the preferred range of concentration of the guest molecule in the blue green host is from 0.01 to 5.0% by mole ratio. Depending the efficiency of fluorescence energy transfer from the host to the guest molecule, choice of guest-host pair, it is understood that the concentration range can be substantially larger than indicated. The upper range of the guest molecule in the host matrix can as high as 10%.

[0039] The electron transport layer 116 of EL device 100 is a metal chelated oxinoid compound, including chelates of oxine itself (also commonly referred to as 8-quinolinol or 8-hydroxyquinoline). Such compounds exhibit both high levels of performance and are readily fabricated in the form of thin films. Exemplary of contemplated oxinoid compounds are those satisfying structural formula (XI).

$$\begin{bmatrix} \begin{bmatrix} Z \\ N \end{bmatrix} \\ Me^{+n} \end{bmatrix}_n \longrightarrow \begin{bmatrix} Z \\ N \\ Me^{+n} \end{bmatrix}_n$$

(XI)

3

wherein

5

10

15

20

25

30

35

Me represents a metal;

n is an integer of from 1 to 3; and

Z independently in each occurrence represents the atoms completing a nucleus having at least two fused aromatic rings.

[0040] From the foregoing it is apparent that the metal can be monovalent, divalent, or trivalent metal. The metal can, for example, be an alkali metal, such as lithium, sodium, or potassium; an alkaline earth metal, such as magnesium or calcium; or an earth metal, such as boron or aluminum. Generally any monovalent, divalent, or trivalent metal known to be a useful chelating metal can be employed.

[0041] Z completes a heterocyclic nucleus containing at least two fused aromatic rings, at least one of which is an azole or azine ring. Additional rings, including both aliphatic and aromatic rings, can be fused with the two required rings, if required. To avoid adding molecular bulk without improving on function the number of ring atoms is preferably maintained at 18 or less.

[0042] Illustrative of useful chelated oxinoid compounds are the following:

Aluminum trisoxine [a.k.a, tris(8-quinolinol)aluminum, (Alq)] Magnesium bisoxine [a.k.a. bis(8-quinolinol)-magnesium] Indium trisoxine [a.k.a., tris(8-quinolinol)indium] Lithum oxine (a.k.a., 8-quinolinol lithium)

[0043] In the organic EL devices of the invention, it is possible to maintain a current density compatible with efficient light emission while employing a relatively low voltage across the electrodes by limiting the total thickness of the organic luminescent medium to less than 10,000-10<sup>-10</sup>m (Angstroms). At a thickness of less than 1 mm an applied voltage of 20 volts results in a field potential of greater than 2 x 105 volts/cm, which is compatible with efficient light emission. An order of magnitude reduction to 100-10<sup>-10</sup>m (Angstroms) in thickness of the organic luminescent medium, allowing further reductions in applied voltage and/or increase in the field potential and hence current density, are well within device construction capabilities.

[0044] The preferred materials for forming the organic luminescent medium are each capable of fabrication in the form of a thin film that is, capable of being fabricated as a continuous layer having a thickness of less than 5000-10-10m (Angstroms). A preferred method for forming the organic luminescent medium is by vacuum vapor deposition. Extremely thin defect free continuous layers can be formed by this method. Specifically, individual layer thicknesses as low as about 50-10-10m (Angstroms) can be constructed while still realizing satisfactory EL device performance. Employing a vacuum vapor deposited porphorinic compound as a hole injecting layer, a film forming aromatic tertiary amine as a hole transporting layer (which can in turn be comprised of a triarylamine layer and a tetraaryldiamine layer), a fluorescent emitting layer comprised of a mixture of a host material and a fluorescent compound, and a chelated oxinoid compound as an electron injecting and transporting layer, individual layer thicknesses in the range of from about 50 to 5000-10-10m (Angstroms) are contemplated, with layer thicknesses in the range of from 100 to 2000-10-10m (angstroms) being preferred. It is generally preferred that the overall thickness of the organic luminescent medium be at least about 1000-10-10m (Angstroms).

[0045] The anode 104 and cathode 108 of the EL device 100 can each take any convenient conventional form. Where it is intended to transmit light from the EL device 100 through the anode, this can be conveniently achieved by

coating a thin conductive layer onto a light transmissive substrate, e.g., a transparent or substantially transparent glass plate or plastic film. In one form the EL device 100 of this invention can follow the historical practice of including a light transmissive anode formed of tin oxide or indium tin oxide coated on a glass plate, as disclosed by Gurnee et al. U.S. Patent No. 3,172,862, Gurnee U.S. Patent No. 3,173,050, Dresner "Double Injection Electroluminescence in Anthracene", RCA Review, Volume 30, pages 322-334, 1969; and Dresner U.S. Patent No. 3,710,167 cited above.

[0046] The EL device 100 of this invention can employ a cathode constructed of any metal, including any high or low work function metal, heretofore taught to be useful for this purpose. Unexpected fabrication, performance, and stability advantages have been realized by forming the cathode of a combination of a low work function metal and at least one other metal. For further disclosure, see commonly assigned U.S. Patent No. 4,885,211 to Tang et al.

#### **EXAMPLES**

10

35

40

45

[0047] The following examples further illustrate the invention.

# 15 SYNTHESIS:

Example 1 - Preparation of 4,4-difluoro-1,3,5,7-tetraphenyl-4-bora-3a,4a,-diaza-s-indacene (G5)

[0048] To a mixture of 3,5-diphenyl-pyrrol-2-carboxaldehyde (0.9 g. 3.6 mmol), which was prepared from 2,4-diphenylpyrrole by Vilsmeier reaction, and 2,4-diphenylpyrrole (0.79 g, 3.6 mmol) in 120 mL of dry dichloromethane was added 0.4 mL of phosphorous oxychloride. The reaction mixture was stirred at room temperature overnight and was then added 2.4 mL of N,N-diisopropylethylamine, followed by addition of 2.4 mL of boron trifluoride etherate. After the reaction mixture was stirred at room temperature for three hours, it was washed with water. The organic layer was separated and then passed through the short silica gel column. After removal of solvents the dark purple 4,4-difluoro-1,3,5,7-tetraphenyl-4-bora-3a,4a,-diaza-s-indacene (1.56 g) was obtained in 87% yield. The pure material used for cell fabrication was obtained by sublimation at 285 °C under 2,67·10<sup>2</sup>N/m<sup>2</sup> (2 Torr).

#### **EL DEVICE FABRICATION AND PERFORMANCE:**

# 30 Example 2 - Fabrication procedure

[0049] An EL device satisfying the requirements of the invention was constructed in the following manner. The device structure has a four organic-layer stack, namely hole-injecting layer, hole transporting layer, luminescent layer, electron-transporting layer.

- a) An indium-tin-oxide coated glass substrate was sequentially ultrasonicated in a commercial detergent, rinsed in deionized water, degreased in toluene vapor and exposed to ultraviolet light and ozone for a few minutes.
- b) A hole injecting layer of copper phthalocyanine (150·10<sup>-10</sup>m (Angstroms)) was then deposited on top of the ITO coated substrate by evaporation from a tantalum boat.
- c) Onto the copper phthalocyanine layer was deposited a hole transporting layer of N,N'-BIS-(1-Naphthyl)-N,N'-Diphenylbenzidine, also evaporated from a tantalum boat.
- d) A luminescent layer of host material (375·10<sup>-10</sup>m (Angstroms)) doped with a guest material was then deposited onto the hole transporting layer. This mixed layer was prepared by co-depositing the two materials from separate tantalum boats. The rates were independently controlled. The typical rate for host material was 5·10<sup>-10</sup>m (Angstroms) per second, and the rate for the guest material was adjusted according to the concentration desired.
- e) A electron transporting layer of Alq (375 Angstroms) was then deposited onto the fluorescent emitting layer.
- f) On top of the Alq layer was deposited a 2000-10<sup>-10</sup>m (Angstroms) cathode formed of a 10:1 atomic ratio of Mg and Ag.
- 50 [0050] The above sequence completed the deposition of the EL device. The device was then hermetically packaged in a dry glove box for protection against ambient environment.

# Example 3- Preparation of white light emitting electroluminescent device

<sup>55</sup> [0051] The EL devices were fabricated according to the procedure of Example 2. Except the fluorescent emitting layer was deposited with host material (H3) doped with various concentration of guest material, 4,4-diffuoro-1,3,5,7-tet-raphenyl-4-bora-3a,4a,-diazasindacene, (G5).

[0052] Table 1 lists the luminance quantum efficiency measured in unit of candela per square meter, CIE color co-

ordinates, and the luminance output under a constant current bias of 20 mA/cm^2.

**[0053]** In the example, the best white emission is from a guest concentration of 0.05% with CIE color coordinates of X = 0.356, y = 0.364, and a luminous sterance of 522 cd/m2 in a current density of 20 mA/cm2.

TABLE 1

The CIE color coordinates and the luminance output under a constant current bias of 20 mA/cm^2.				
Conc. of dopant	0.0%	0.05%	0.1%	0.25%
Cd/m^2	495	522	445	365
Cie x	.2069	.3555	.4115	.4640
Cie y	.3352	.3640	.3656	.3827

[0054] This result demonstrates the production of white light from single layer luminescent layer containing a red light emitting material uniformly dispersed in the host material in accordance with the present invention.

# **PARTS LIST**

# [0055]

5

10

15

20 100 electroluminescent device

102 glass substrate

104 anode

106 organic electroluminescent medium

108 cathode

110 hole injection layer

112 hole transport layer

114 luminescent layer

116 electron transport layer

120 power source

30 122 conductors

124 conductors

130 hole

132 arrows

140 electrons

35 142 arrows

### Claims

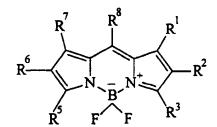
40

45

50

55

1. An electroluminescent device (100) comprising, in sequence, an anode (104), a hole-injection layer ((110), a hole transport layer (112), a luminescent layer (114), an electron-transporting layer (116), and a cathode (108), the luminescent layer (114) including a host organic material uniformly doped with a guest component having the formula:



wherein R<sup>1</sup>-R<sup>8</sup>, which may be the same or different, are hydrogen, halogen, or alkyl, alkoxy, alkenyl, cycloalkyl, arylalkyl, acyl, wherein the alkyl portions each containing fewer than 24 carbons, or aryl heteroaryl, alone or in

combination, wherein the host material comprises a 8-quinolinol Aluminum complex of the formula:

$$(R^S \longrightarrow Q \longrightarrow AI \longrightarrow O \longrightarrow L$$

wherein Q in each occurrence represents a substituted 8-quinolinolato ligand, R<sup>S</sup> represents an 8-quinolinolato ring substituent chosen to block sterically the attachment of more than two substituted 8-quinolinolato ligands to the aluminum atoms, O-L is a phenolato ligand, and L is a hydrocarbon group that includes an aryl moiety.

2. The invention according to claim 1 wherein the guest component produces orange-red fluorescence in dilute solution with a high quantum efficiency.

## Patentansprüche

5

10

15

20

25

30

35

40

45

50

55

Ein elektrolumineszentes Bauteil (100), das aufeinanderfolgend aufweist: eine Anode (104), eine Löcherinjektionsschicht (110), eine Löchertransportschicht (112), eine Lumineszenzschicht (114), eine Elektronentransportschicht (116) und eine Kathode, wobei die Lumineszenzschicht (114) ein organisches Gästmaterial aufweist, das
gleichmäßig dotiert ist mit einer Gastkomponente mit der Formel:

wobei R¹-R³, die gleich sein können oder unterschiedlich, Wasserstoff, ein Halogen oder ein Alkyl, ein Alkoxy, Alkenyl, Cycloalkyl, Arylalkyl, Acyl sind, wobei die Alkylteile jeweils weniger als 24 Kohlenstoffe beinhalten, oder Arylheteroaryl allein oder in Kombination, wobei das Gastmaterial einen 8-Quinollnol-Aluminiumkomplex aufweist von der Formel:

$$(R^S - Q - AI - O - L$$

wobei Q jedesmal, wenn es vorkommt, einen substituierten 8-Quinolinolatoliganden repräsentiert, wobei R<sup>S</sup> einen 8-Quinolinolatoringsubstituenten repräsentiert ausgewählt für ein sterisches bzw. räumliches Blockieren der Anbringung von mehr als zwei substituierten 8-Quinolinolatoliganden an die Aluminiumatome, wobei O-L ein Phenolatoligand ist und L eine Hydrocarbongruppe ist, die einen Arylteil enthält.

2. Erfindung gemäß Anspruch 1, wobei die Gastkomponente eine orangerote Fluoreszenz in verdünnter Lösung mit einer hohen Quanteneffizienz erzeugt.

#### Revendications

1. Dispositif électroluminescent (100) comprenant, dans l'ordre indiqué, une anode (104), une couche d'injection de trous (110), une couche de transport de trous (112), une couche luminescente (114), une couche de transport d'électrons (116) et une cathode (108), la couche luminescente (114) comprenant un matériau organique hôte uniformément dopé au moyen d'un composant hôte représenté par la formule :

$$R^{6} \xrightarrow{R^{7}} R^{8} \xrightarrow{R^{1}} R^{1}$$

$$R^{6} \xrightarrow{R^{5}} F \xrightarrow{R^{5}} R^{3}$$

10

5

où R1-R8, qui peuvent être identiques ou différents, sont l'hydrogène, l'halogène ou un groupe alkyle, alcoxy, alcényle, cycloalkyle, arylalkyle, acyle, où les portions alkyle contiennent chacune moins de 24 atomes de carbone ou un groupe aryl hétéroaryle, seuls ou en combinaison, où le matériau hôte comprend un complexe d'aluminium de type 8-quinolinol représenté par la formule :

20

15

$$(R^S - Q)$$
  $\rightarrow Al$   $-O$   $-L$ 

où Q dans chaque cas représente un ligand de type 8-quinolinolato substitué, Rs représente un substituant cyclique de type 8-quinolinolato choisi de manière à bloquer stériquement le rattachement de plus de 2 ligands 8-quinolinolato substitués aux atomes d'aluminium, O-L est un ligand phénolato et L est un groupe hydrocarboné comprenant un radical aryle.

2. Invention selon la revendication 1, dans laquelle le composant hôte produit une fluorescence orange-rouge dans

une solution diluée avec un rendement quantique élevé.

30

25

35

40

45

50

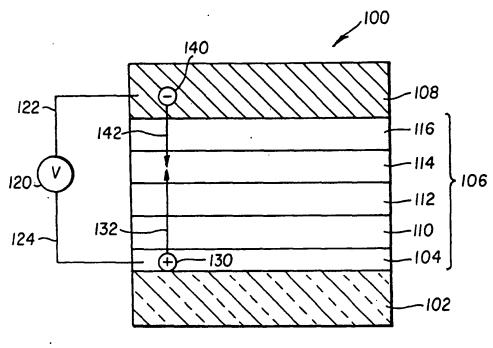
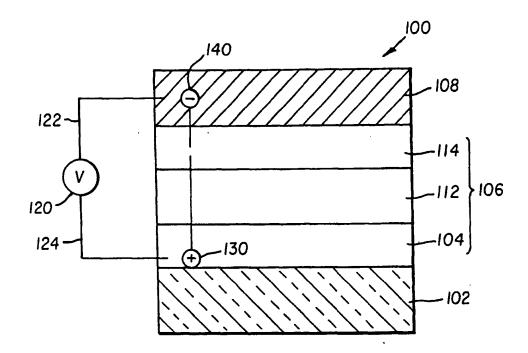


FIG. I



. FIG. 2

This Page Blank (uspto)